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ULTRAFAST PROCESSES AND SPECTROSCOPY WITH FREE ELECTRON 1/1  
LASERS(U) PRINCETON UNIV NJ DEPT OF ELECTRICAL  
ENGINEERING AND COMPUTER SCIENCE P M FRUCHET APR 88

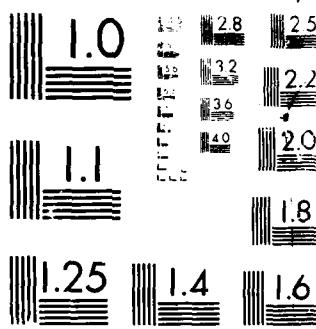
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19. ABSTRACT (Continue on reverse if necessary and identify by block number) Femtosecond laser spectroscopy has been used to study carrier relaxation times in amorphous silicon. We find a relaxation time of 1 picosecond above the mobility edge and a relaxation time of 10 picoseconds in the bandtail states, after which temperature effects dominate the optical properties. Theoretical modeling of femtosecond spectroscopic measurements has also helped define what is measurable and what is not.			
Picosecond time-resolved reflectivity measurements have been performed during laser-induced phase transitions. The dielectric function of molten Si has been measured and superheating in the liquid phase has been observed to last at least 10 picoseconds.			
Work continues in both areas. We expect to expand the experimental program to other wavelengths thanks to the free electron laser.			
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# Annual Report for contract N00014-86-K-0711

## Ultrafast processes and spectroscopy with free electron lasers

prepared by

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The goal of this project is to uncover the electronic processes in disordered materials. Understanding these processes is the key to understanding the physics in materials such as amorphous semiconductors or liquid semiconductors. In turn, understanding the physics of these materials will lead to novel devices or improved device operation and will impact on our models for other materials.

We have concentrated on time-resolved measurements of the optical properties of silicon, both amorphous and liquid. We have used the methods of picosecond and femtosecond time-resolved reflectivity and transmission. The experiments have been performed with conventional lasers but will be moved to our laboratory located at Stanford University when the FEL becomes operational.

### Results

#### a. Amorphous silicon

Our previous work had been exclusively experimental and had shown that the accepted interpretation of picosecond photoinduced absorption in a-Si:H was at least in part incorrect [1]. We have now performed additional experiments which have allowed us to refine our ideas and we have also obtained some interesting theoretical results. Those theoretical results [2] indicate clearly that the interpretation of pump-and-probe experiments differ for crystalline and amorphous materials. The main reason is that momentum conservation rules which exist for optical transitions in crystalline materials are relaxed in disordered materials. We have shown that carrier-carrier scattering cannot be measured from conventional experiments, and that carrier-phonon scattering, although measurable, leads to much smaller effects than with crystalline materials. In particular, no hole burning is observable.

In our new experiments, we have now confirmed that the relaxation of hot carriers above the mobility edge takes 1 picosecond and that at later time, the sample temperature rise produced by the energy lost by the carriers plays an important role. In fact, the temperature appears to be the best probe of the energy relaxation in the bandtail states, since we have found that the optical

cross-section for intraband absorption from (localized) bandtail states to extended states is much smaller than that for transitions between extended states. After 10 picoseconds, the relaxation processes slow down dramatically, in agreement with previous results that indicated bandtail relaxation times in the 1 nanosecond range. This is consistent both with attainment of quasi-equilibrium and with the increased difficulty of relaxation from relatively deep states. The results are amplified in our very recent publications [3-6].

### b. Liquid silicon

We have investigated the properties of liquid silicon produced by high power laser illumination. This study is to be considered in the context of laser-induced phase transitions and of laser-induced damage, two phenomena which are possible with ultrashort high power FEL pulses. In recent work [7], we had demonstrated a powerful technique to measure the real and imaginary parts of the dielectric function of any solid during and after picosecond laser-induced phase transition. We have now applied this technique to the determination of the dielectric function of liquid silicon close to the melting temperature [8,9]. We have also shown for the first time that laser-produced liquid phases could exceed the boiling temperature for many picoseconds without undergoing the phase transition [9,10]. This new result should open new avenues of research in non-equilibrium thermodynamics.

### Future work

The work on picosecond and femtosecond spectroscopy of amorphous semiconductors is continuing vigorously. We have planned new series of experiments, in samples of various bandgaps and at variable temperatures. We are also planning to repeat some experiments with 30 femtoseconds time resolution (instead of 100 fs), which we believe will increase our understanding of the relaxation processes. We intend to use the FEL beam to perform much-needed spectroscopy in the infrared (beyond 1500 nm, where subpicosecond laser sources are not available). Further theoretical work is also planned, especially with regard to the position of the mobility edge. The work on non-equilibrium laser-induced phase transitions will resume when the FEL is operational. This work could impact on the optical damage often observed in FEL cavities.

### Publications

1. P.M. Fauchet, D. Hulin, A. Migus, A. Antonetti, J. Kolodzey, and S. Wagner, "Initial stages of trapping in a-Si:H observed by femtosecond spectroscopy," Phys. Rev. Lett. **57**, 2438 (1986).
2. P.M. Fauchet and K. Gzara, "Determination of carrier-carrier and carrier-phonon relaxation times from ultrafast photoinduced absorption in amorphous semiconductors," submitted for publication.

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3. P.M. Fauchet, D. Hulin, A. Migus, A. Angonetti, J.P. Conde and S. Wagner, "Femtosecond spectroscopy in amorphous silicon and silicon-germanium alloys," *J. Non-Cryst. Solids* **97 & 98**, 145 (1987).
4. P.M. Fauchet et al., "Hot carrier dynamics in amorphous semiconductors," to be published in SPIE Proc.
5. "Carrier transport in amorphous semiconductors," invited talk at the American Physical Society Meeting, New Orleans, March 1988.
6. P.M. Fauchet et al., in preparation.
7. K.D. Li and P.M. Fauchet, "Picosecond determination of the dielectric function of liquid silicon at 1064 nm," *Solid State Commun.* **61**, 207 (1987).
8. K.D. Li and P.M. Fauchet, "Drude parameters of liquid silicon at the melting temperature," *Appl. Phys. Lett.* **51**, 1747 (1987).
9. P.M. Fauchet and K.D. Li, "The dielectric function of laser-produced molten silicon," *J. Non-Cryst. Solids*, **97 & 98**, 1267 (1987).
10. P.M. Fauchet and K.D. Li, "Picosecond laser induced melting: the dielectric function of molten Si and superheating in the liquid phase," to be published in the *Proceedings of the Materials Research Society*.

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